Multi-conical second harmonic waves via nonlinear diffractions in circularly poled nonlinear media

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ABSTRACT

We investigate nonlinear diffraction (NLD) of laser radiation in circularly poled nonlinear quadratic crystal for the case of single and two fundamental pump beams. We show that **single pump beam** excitation (10 ps @ 1.053 μm) along Z axis of circularly poled structure (with period 7.5 μm) leads to the second harmonic signal being emitted in a form of multiple low order cones (rings) and one strong external SH cone (ring) defined by the longitudinal phase matching conditions. We study a dependence of the NLD pattern as a function of the incidence angle of the pump. We demonstrate that **two noncollinear pump beams** intersecting exactly in the center of the structure results in a new type nonlinear diffraction, which does not have an analogue in linear optics. It features a set of nonlinearly diffracted beams originating from each individual pump accompanied by the set of additional diffraction rings which originate from photons coming from both pumps. The corresponding phase matching conditions responsible for the observed NLD effects are discussed. The observed effects represent nonlinear generalization of optical diffraction in linear media and we believe can find possible applications in second harmonic optical microscopy.

Keywords: nonlinear optics, second harmonic generation, diffraction

1. INTRODUCTION

Wavelength conversion of light via second harmonic generation (SHG) is one of the most commonly known nonlinear processes. Since its discovery in 1961, this process has revolutionized the optical research and industry, having a vast impact on the development of lasers, laser spectroscopy, and microscopy. Naturally available nonlinear materials cannot meet today's demands for applications of SHG. This has driven the development of new crystalline materials with suitable properties and micro-engineering of the ferroelectric structure of existing materials to fine tune them. The new engineered nonlinear structures exhibit one - or two-dimensional spatial modulation of the sign of nonlinearity realized by means of electrical poling. The most frequently used structures include structures with either constant, chirped or phase reversed period, super lattices with Fibonacci and other generalized Fibonacci sequences. The two dimensional pattern modulation of nonlinearity include rectangular, hexagonal, quasiperiodic, and partially or totally disordered paterns. Such two dimensional structures enable also generation of nontraditional SH waves like Bessel [1] and toroidal waves [2]. The emission of such waves is determined by the transverse phase matching condition and hence can be easily explained implementing the concept of nonlinear diffraction [3]. In most of the experiments with periodically poled structures the pump is directed perpendicularly or close to perpendicular direction with respect to axis Z. In our previous publication [1] we investigated for the first time the interaction for pumping along Z axis of annularly poled structure of Stoichiometric Lithium Tantalate with femtosecond Ti:Sapphire laser operating at 835 nm.

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15th International School on Quantum Electronics: Laser Physics and Applications, edited by Tanja Dreischuh
Elena Taskova, Ekaterina Borisova, Alexander Serafetinides, Proceedings of SPIE Vol. 7027, 70271T
· doi: 10.1117/12.822542

So far the nonlinear diffraction (NLD) along Z axis, has been demonstrated only in single beam experiments[1,4] The effect of diffraction in linear optics is usually observed in the propagation of linear waves in a medium with a periodic change of the optical refractive index[5]. For certain angles of incidence, the partially refracted waves add coherently and form strong beams propagating outside the sample at the angles $\beta_m = \arcsin(m\lambda/\Lambda)$ where λ is the wavelength, Λ is the modulation period, and m is an integer number representing the diffraction order. Importantly, similar diffraction can also occur in media with a spatially homogeneous optical refractive index but periodically varying nonlinear properties. For example, the nonlinear Bragg diffraction, was first discussed by Freund [3] and it was observed experimentally in naturally laminated crystals exhibiting non-regularity and dispersion of nonlinear domains[6,7]. Recently, nonlinear diffraction has also been reported in the second- and third-harmonic generation in the presence of a transient optically-induced 1D grating[8]. Nonlinear diffraction is observed also in photonic crystals with simultaneous modulation of both linear and nonlinear optical properties [9,10]. In addition, nonlinear diffraction with multi-order generation of SH is observed in one dimensional gratings formed by a series of waveguides made by proton exchange [11].

We have recently studied nonlinear diffraction along Z direction from circularly poled Stoichiometric Lithium Tantalate crystal with the fundamental beam (150 fs pulse at 835 nm laser wavelength from Ti:Sapphire laser) propagating along Z axis [1]. In the recently published paper Shutov at al [4] reported on second harmonic in 1D periodically poled LiNbO3 crystal using also femtosecond Ti:Sapphire laser beam incident at the sample along Z axis. The authors observed two pair of spots of phase matched second harmonic (SH) signal located symmetrically on both sides of the pump, which they believe are result of 41 order transverse phase matching within the QPM structure. However, their theoretically predicted angles of the emission of the SH differed by few degrees from the experimental measurements. As we show below in such phase matched SH generation one has to rely on the longitudinal phase matching conditions in order to predict correctly the direction of generated second harmonic.

In this paper we report on theoretical and experimental studies of conical second-harmonic generation from a 2D nonlinear photonic structure fabricated in a periodically poled stoichiometric lithium tantalate (SLT) crystal with annular periodic modulation of the second-order nonlinearityp, excited by picosecond laser pulses at 1053 nm.

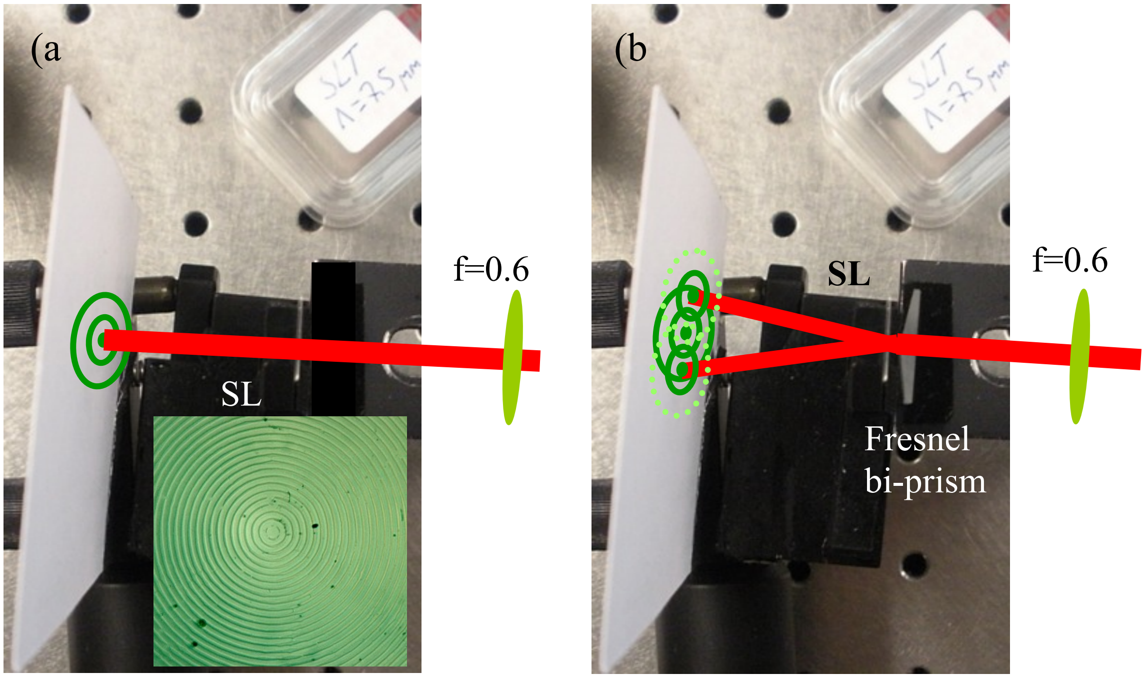


Fig. 1 Schematic of the experiment for observation of nonlinear diffraction in circularly poled stoichiometric lithium tantalite: a) single fundamental beam experiment; b) two fundamental beams experiment

2. EXPERIMENTAL RESULTS AND DISCUSSIONS

We observe that the transverse illumination of a radially periodic structure by one or two fundamental beams exactly in the center of the structure leads to multiple conical emission of SH waves outside the sample.

2.1 Single beam excitation

The schematic of the experiment with a single fundamental beam is shown in Fig.(1a). The incident beam is tightly focused in the central part of the annularly periodically poled SLT crystal. The second harmonic signal is observed on the screen located behind the sample. It appears that SH waves form a set of rings in the far-field zone, and the angles of their propagation satisfy the well-known diffraction equation in linear optics i.e., they are determined by a ratio of the SH wavelength to the modulation period of the nonlinearity. We showed theoretically [12] that in such an annular quasi-phase-matched (QPM) structure the SH light field inside the sample is composed of Bessel beams of different widths and orders, so that the diffraction rings recorded are in fact the far-field images of the SH Bessel beams generated inside the nonlinear crystal.

The stoichiometric lithium tantalate (SLT) sample is described in Ref. [13.] It is 0.49 mm thick, with a QPM period of 7.5 μm. The microphotography of +Z surface of the sample is shown in the inset of Fig. 1(a). To observe the effect of the generation of conical SH waves, we employed an experimental setup that consists picosecond laser at 1053 nm with a regenerative amplifier. It delivers 10 ps pulses with repetition rate of 20 Hz and output pulses energy close to 1 mJ.

In Fig. 2(a) we show the recorded nonlinear diffraction pattern obtained with single beam excitation at normal incidence (along axis Z). The image in Fig.2(b) corresponds to the situation when the fundamental beam is directed at angle 10° with respect to crystallographic Z-axis while the screen remains normal to the direction of the fundamental beam.

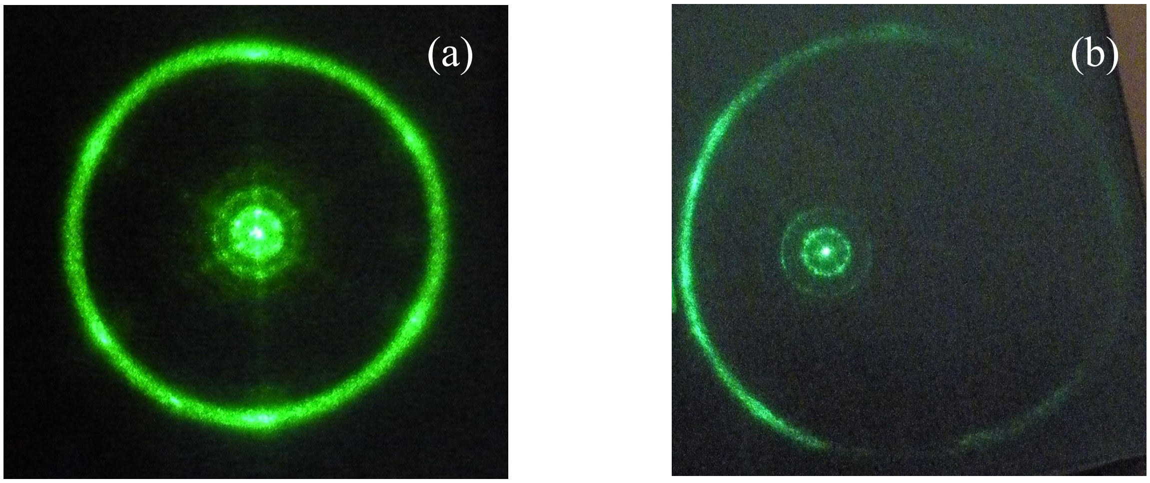


Fig. 2 SHG in SLT with 1053 nm pumping: a) normal incidence - exactly along Z axis; b) 10° angle incidence with respect to axis Z

The internal rings seen in Fig. 2(a,b) are results of the nonlinear diffraction (NLD) of the fundamental beam on the circular $\chi^{(2)}$ grating [1] and for this reason we will call them NLD rings. Each ring correspond to a conical wave with the characteristic angle defined by the relation

$$\alpha_m = \arcsin\left(m \frac{G_o}{k_2} \right), \tag{1}$$

in case of normal incidence of the fundamental beam (FB) to input face of the sample (FB is along Z). Eq.(1) is obtained as a result of fulfilling the transverse PM condition $k_2 \sin\alpha_m - mG_o = 0$. In this case and for the normal incidence the rings (cones) are centered around the axis Z and the fundamental beam. Knowing that the period of the annular grating is 7.5 μm we obtain for the first two NLD rings following conical (external) angles 4.02^o and 8.07^o. The experimentally measured values are 4.0^o and 8.1^o, respectively and coincide perfectly with the predictions.

In case of oblique incidence of the FB with respect to the axis Z the NLD rings correspond to the following conical angles:

$$\alpha_m = \arcsin\left(m\frac{G_o}{k_2}\cos\gamma \right) \tag{2}$$

where γ is the angle of incidence. Λ and m has been defined in the Introduction section. Eq. (2) can be easily derived

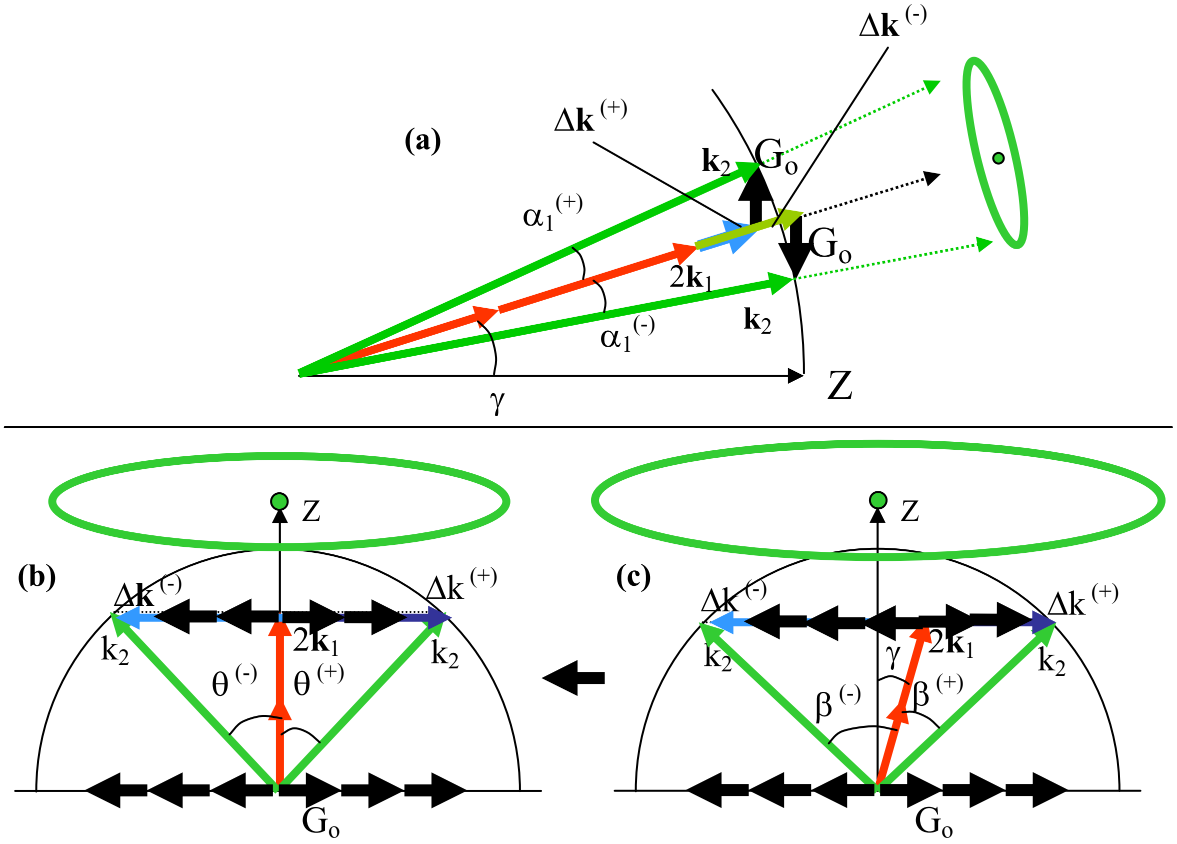


Fig. 3 Phase matching diagrams: a) for observation of NLD rings at angle excitation with respect to axis Z. b) and c) for observation of the PM ring with normal and oblique excitation respectively

using the phase matching diagram shown in Fig. 3(a). As can be seen it predicts that the internal angles are

$$\alpha_m^{(+)} = -\alpha_m^{(-)}. \tag{3}$$

independently of the value of the incidence angle γ. That means the internal NLD rings remain symmetrically situated with respect to the pump but not with respect to the Z axis. This is confirmed by the experiment as seen in Fig.2(b). On the screen the shape of the rings for relatively big angles γ will be distorted due to the fact that light refracts at the output surface of the crystal. Additionally for $\gamma \neq 0$ the shapes of the rings on the screen depend also on the fact the screen is perpendicular to the pump or to the axis z. From Eq. (2) follows that the internal angles of the NLD cones decrease with increasing of the angle γ.

We have to note the PM diagrams shown in Fig.3(a) assumes that the mismatch vector Δk are directed along the direction of the pump. The other option (also discussed in the literature) is to assume the direction of vector Δk to be perpendicular to the grating vector G_o. This approach leads internal rings to be strongly asymmetrically situated with respect to the pump that is in striking contradiction with the experimental observations.

Let us now consider the behavior of the strong large diameter SH ring (cone). When the FB propagates along the Z axis, the this ring is symmetric with respect to the FB and the axis Z. Its internal cone angle is defined by the longitudinal phase matching (LPM) condition [1, 14,15]

$$\cos\theta^{(\pm)} = \frac{2k_1}{k_2} \tag{4}$$

illustrated in Fig. 3(b). The angles measured from both sides of the pump and axis Z are the same. Using the values of index of refraction given in Ref [16] we calculated the internal LPM-SH cone angles for the interaction OO-E to be $\theta^{(\pm)}[O_1O_1 - O_2] = 14.37^o$ and $\theta^{(\pm)}[O_1O_1 - E_2] = 14.27^o$, where “O” and “E” denotes ordinary and extraordinary beam, respectively. These values correspond to 33.21^o and 32.91^o external SH cone angles. As they are very close, we do not resolve them in the experiment so only a single ring is visible on the screen. The predicted ratio for the internal angles for normal incidence is

$$R_{\text{int}} = \frac{\theta}{\alpha_1} = \frac{\arccos(2k_1/k_2)}{\arcsin(G_o/k_2)} \quad , \tag{5}$$

that for the external angles became:

$$R_{ext} = \frac{\arcsin\{n_{2o}.\sin[\arccos(2k_1/k_2)]\}}{\arcsin(\lambda_2/\Lambda)}. \tag{6}$$

In our experimental conditions, we find that theoretically predicted and experimentally measured values of R_{ext} are 8.2 and 7.8 respectively. Then the experimental LPM external angle is 31.2^o compared with the theoretical value 33^o. The difference may be is caused by the lack of the “good” Sellmeier equations for the ordinary index of refraction of SLT.

When the nonlinear crystal is rotated at angle $\gamma \neq 0$ with respect to the fundamental beam the emitted LPM ring is situated asymmetrically with respect to the directions of the FB. However, the ring remains centered with respect of the Z axis. For the conical angle is valid the relation $\theta_\gamma^{(-)} = \theta_\gamma^{(+)}$ for any γ. The phase matching conditions for this case are illustrated in Fig.3(c). The angles from both sides of the pump are now different

$$\beta^{(-)} > \beta^{(+)}, \tag{7}$$

but the LPM emission retains its circular shape with the center of the cone coinciding with the Z-axis. The conical angle is now determined by the following relation [15, 17]

$$\cos\theta_{\gamma}^{(\pm)} = \frac{2k_1}{k_2}\cos\gamma\,. \tag{8}$$

Therefore, if viewed on a screen perpendicular to the Z axis the ring diameter becomes bigger. This was verified and confirmed in the experiment. In addition to that, since (i) Δk (the mismatch parameter) varies with the azimuthal angle and (ii) QPM order also varies with the azimuthal angle , so does the intensity of the emitted SH signal. If the screen remains perpendicular to the fundamental beam than we record cone section – an ellipse as shown in Fig. 2(b). The bigger is the angle γ, the bigger is the eccentricity of the SH ellipse. It can be shown that eccentricity is related to the external cone angle θ_{ext} and the angular position of the crystal ϕ (with respect to the pump) by simple relation:

$$\varepsilon = \sin\phi / \cos\theta_{ext}\,. \tag{9}$$

In the example illustrated in Fig. 2(b) which was obtained by tilting of the SLT crystal by 10° the measured value of eccentricity was $\varepsilon = 0.18$, which is very close to theoretically predicted value of $\varepsilon = 0.21$.

The formulae (4) and (8) have exactly the same form as those that characterize the conical waves in random nonlinear crystals with antiparallel ferroelectric domains [14, 15, 18, 19]. We carefully verified that the size of the PM ring obeys the formula Eq.(4) which represents just longitudinal phase matching (LPM) condition of the general vectorial form

$$\mathbf{k}_2 - 2\mathbf{k}_1 + m\mathbf{G}_o + \mathbf{\Delta k} = 0\,. \tag{10}$$

The conclusion that we reached is that the cone angle of the strong SH external ring is defined exactly by the **LPM** condition and the intensity of the ring depends on the magnitude of transversal phase-mismatch Δk_{TM} (the smaller is Δk_{TM} the higher is the intensity of the SH-LPM ring) :

$$\Delta k_{TM} = k_2 \sin\theta - mG_o\,. \tag{11}$$

If G_o is small then several orders contribute to the intensity of the PM ring.

Table 1. Comparison of properties of the two types of the SH rings observed

TYPE OF THE RINGS	NORMAL INCIDENCE	OBLIQUE INCIDENCE
TPM RINGS	**centered on Z** **centered on FB**	**not centered on Z** **centered on FB** **decrease of the diameter***
LPM RING	**centered on Z** **centered on FB**	**centered on Z** **not centered on FB** **increase of the diameter***

*) on screen perpendicular to axis Z; with respect to normal case

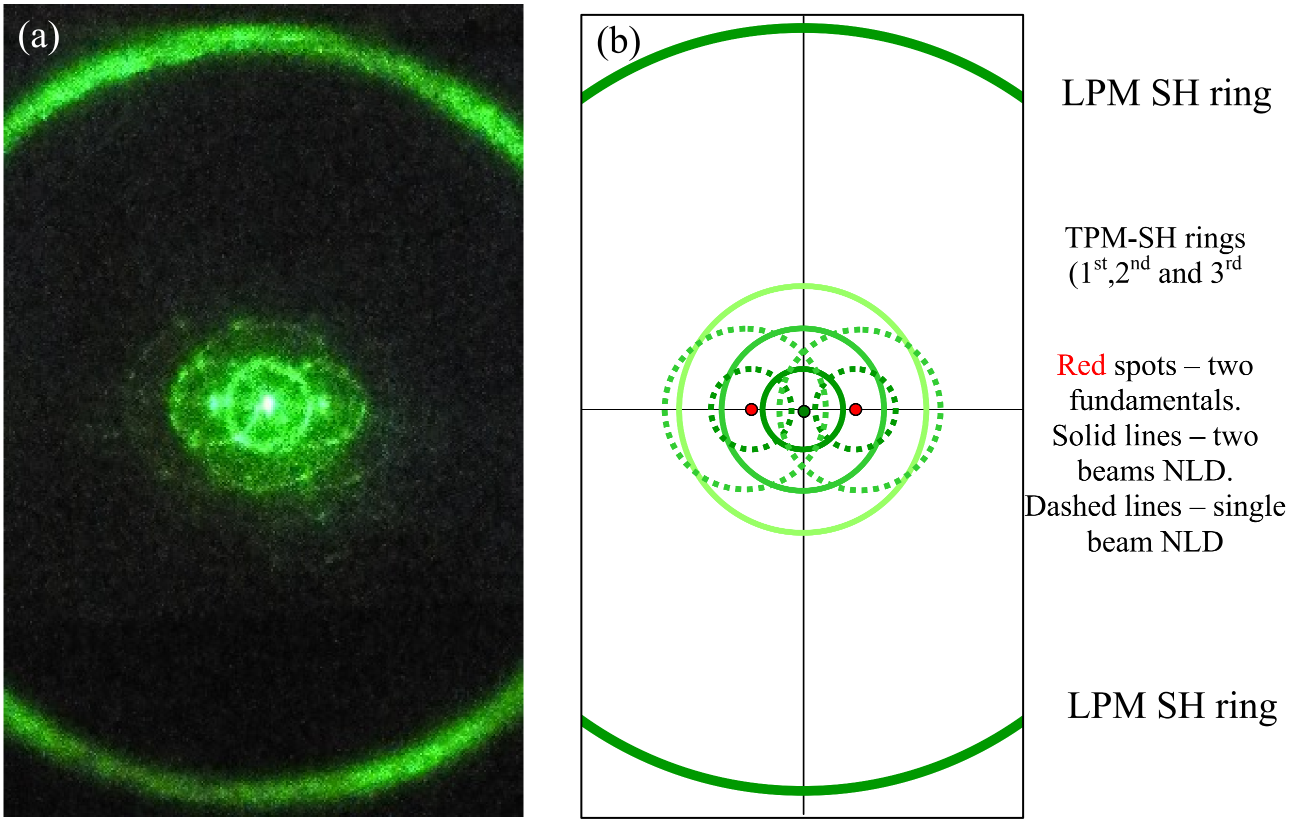
We see the behavior of the TPM rings and the LPM ring are drastically different. The properties of the two types of rings are summarized in Table 1.

Concerning the polarization properties of the rings and the azimuthal modulation of the intensity of the rings we do not discuss them here in details since they are the same as reported in [1,20]

2.2 Two beam excitation

The schematic of second harmonic generation with two beam excitation is shown in Fig.1(b). To create the two beams we use biprism of Fresnel made of BK7 glass with the roof angle of 160°. This results in the formation of two fundamental beams (1053 nm) intersecting at an angle of $2\gamma = 10.34^{\circ}$. In such experimental conditions, we recorded and investigated new type nonlinear diffraction, having no analogue in linear optics, that is observed with the two non-collinear beams crossing exactly in the center of the annularly poled structure . The resulting SH nonlinear diffraction pattern is shown in Fig. 4(a). The two side rings [see the center in Fig. 4(a) and Fig. 5 (right)] are result from the 1st order single beam NLD (the left and right ring). On the other hand, the central rings (both 1st and 2nd orders are visible) are a result of two-beam NLD and they appear only if the two fundamental beams overlap in time and space in the center of the structure. The cone angle of the 1st order side rings are defined by the relation Eq. (2). The cone angles of the central rings do not depend on the biprism angle and are defined by Eq. (1), that was verified in the experiment. The reason for that is that these cone angles are determined by the exact fulfilling of the transverse PM condition

$$k_2 \sin\alpha_m - mG_o = 0. \tag{12}$$



experiment prediction

Fig. 4 a) Experimentally observed SHG with two noncollinear pumps along Z axis of circularly poled SLT. b) prediction based on known angle between fundamental pumps and the period of poling.

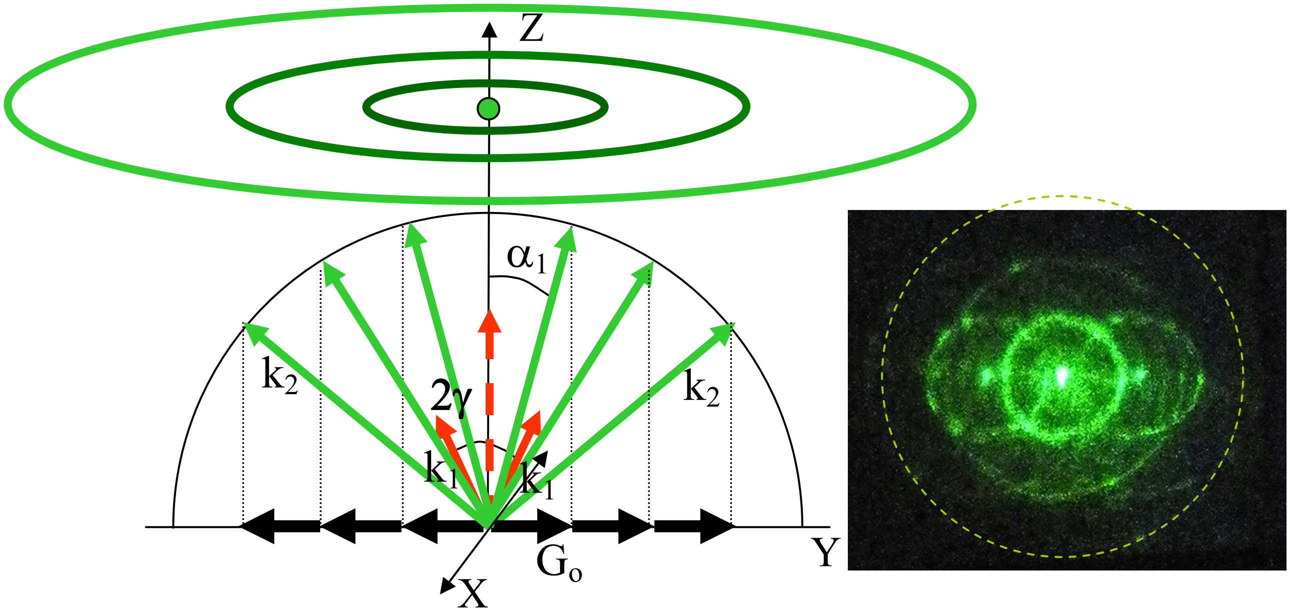


Fig. 5 Left: Phase matching diagram for observation of two beam nonlinear diffraction. 2γ is the angle of noncollinearity situated in the plane perpendicular to the plane of the drawing. Right: TPM SH rings from two noncollinear fundamental beams. The dash line indicate the position of 3rd order SH ring due to two FB nonlinear mixing.

The central spot is a non-phase matched SHG resulting from the noncollinear mixing of the two fundamental beams. The two side spots are nonphase matched SH signals generated by each individual FB. The phase matching diagram for the two beam nolinear diffraction is shown in Fig. 5(left). As seen in Fig. 4 and Fig.5 (right) the strongest is the first order ring. Every next higher-order ring is less intensive, since it requires higher order phase matching. We have to point

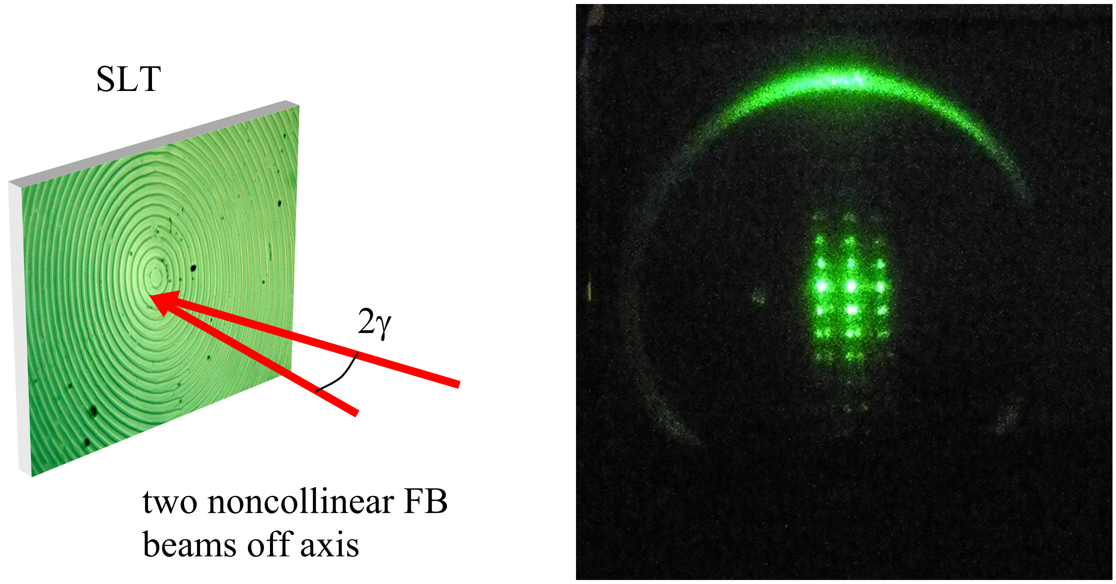


Fig. 6 Higher order SH nonlinear diffraction obtained by off-axis excitation of the sample in vertical direction with two noncollinear pumps. Left- the sample and the two beams. Right the image of the obtained diffraction pattern. The two sides array of spots are from the individual pump beams, while the middle line is a result of two pump beam mixing. The vertical period is defined by the period of the annular structure, while the horizontal distance between the lines of spot is defined by the bi-prism angle.

out that these NLD rings result from the fulfillment of only transverse PM condition.

To the intensity of the stronger, large diameter ring called LPM ring in Fig.4 in fact contribute three overlapping rings: two from the two individual pumps and the third one due to mixing of the two fundamental pumps. All these three cones have the same cone angle defined by the formula Eq.(8) and overlap in case of normal fundamental beam incidence. However they can be perfectly resolved if the sample is tilted strongly with respect to the propagation direction of the fundamental beams.

Fig. 6 illustrates higher order SH nonlinear diffraction which has been obtained by off-axis excitation of the sample (i.e. shift in vertical direction). In this particular case, the structure behaves as a one- dimensional $\chi^{(2)}$ grating. Emission spots corresponding up to 6th order of the two-beam NLD are visible. The two sides array of spots are from the individual pump beams, while the middle line is a result of two beam TPM diffraction. The vertical period is defined by the period of the annular structure, while the horizontal distance between the lines of spot is defined by the biprism angle.

3. CONCLUSION

In conclusion the observed effects contribute to the further development of new branch of Nonlinear Optics – along Z Nonlinear Optics that started with investigation of random SBN crystal [14,15,18] and now is observed in 2D nonlinear photonic structures with periodical modulation of second order nonlinearity. Experiments with other types 2D structures are in progress. The observed effect is nonlinear generalization of diffraction in linear media and can find possible applications in SH optical microscopy.

ACKNOWLEDGMENTS

The work has been supported by the Australian Research Council and the Israeli Science Foundation, grant no. 960/05. We are gratefull for the assistance of D. Kasimov, A. Bruner, P. Shaier, and D. Eger in producing the SLT sample. Solomon Saltiel thanks the Nonlinear Physics Centre and the Laser Physics Centre of the Australian National University for a warm hospitality and support.

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